# Tropical Composition and Climate Coupling (TC³) Experiment

## *Draft White Paper*

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# Tropical Composition and Climate Coupling (TC$^3$) Experiment

## 1. Overview

The composition of air transported from the upper tropical troposphere defines the chemical boundary condition for the global stratosphere and for the upper troposphere in the subtropics. These boundary conditions influence the composition and climate of the entire middle atmosphere. The recovery of stratospheric ozone due to the ban on CFCs will be influenced by how these boundary conditions may change in the coming decades. For example, an increase in the abundance of stratospheric water will, all else being equal, lead to a decrease in the concentration of ozone. In a broader sense, understanding the physical processes that control water in the tropical upper troposphere is critical for assessing how surface temperatures will be affected by rising concentrations of greenhouse gases. The upper tropical troposphere is perhaps the most important region for understanding how stratospheric composition and climate are coupled.

The Tropical Composition and Climate Coupling Experiment (TC$^3$) will obtain, over several years, a comprehensive set of observations to investigate the physics and chemistry in the region of the tropical tropopause. A two-site Pacific aircraft campaign is proposed with flights from Guam (13.4°N, 144.4°E) and Hawaii (21.3°N, 157.9°W) coupled with balloon-borne observations from a yet-to-be-determined tropical site. The science goals of TC$^3$ will benefit directly from the availability of AURA measurements that will place the local observations within a global and long-term context. In return, TC$^3$ is designed to maximize the availability of co-located profiles for AURA validation.

TC$^3$ will address the following questions:

- *What are the physical mechanisms that control the humidity of the stratosphere?*

  The increase of greenhouse gases is expected to alter the amount of water entering the stratosphere. Such changes can profoundly affect stratospheric chemistry. At mid-latitudes, $H_2O$ supplies the $HO_x$ radicals that directly catalyze removal of $O_3$. Higher levels of $H_2O$ exacerbate $O_3$ loss by promoting more efficient heterogeneous chlorine activation at mid-latitudes and in polar regions during winter. The ability to understand how stratospheric water may change in the

coming decades is limited, however, by our uncertainty about the precise mechanisms that dehydrate air entering the stratosphere.

- *What is the chemical fate of short-lived compounds transported into the upper troposphere?*

    The chemical properties of tropical upper troposphere define the chemical boundary condition for the stratosphere. Despite a decade of tremendous progress in tropospheric and stratospheric chemistry, the photochemistry of the upper troposphere remains poorly constrained. This uncertainty limits our ability to understand the fates of numerous compounds (and their degradation products) transported into the near tropopause region. Such knowledge is required if we are to understand the impact of climate change on the stratosphere in general and ozone in particular.

- *What mechanisms maintain the humidity of the tropics and subtropics?*

    The response of the hydrological cycle to changes in the concentration of greenhouse gases is perhaps the single most important source of uncertainty in predicting future changes to Earth's climate and composition. The small amounts of water vapor in the upper troposphere (UT) exert enormous leverage on Earth's radiative balance. Of particular importance is the moisture in the subtropical regions. These dry regions have a large cooling effect on the whole tropics. Understanding the mechanism that controls the humidity of the subtropics is key to determining the nature of the water vapor feedback on climate.

These three key questions are related by a common observational strategy. TC$^3$ will provide a large suite of in situ and remote measurements of tracers of atmospheric motion and chemistry. The same tracers can be used to test the series of hypotheses outlined below regarding the physical and chemical mechanisms that control the distribution of ozone, water vapor and other important trace constituents of the lower stratosphere (LS) and upper troposphere (UT). AURA (and other satellite) observations within the UT/LS will provide information on the spatial, seasonal, and interannual variations while the two-site Pacific aircraft campaign is designed to focus on smaller spatial scale process studies. Aircraft and balloon observations obtained during northern winter and summer will enable examination of important seasonal differences in transport and chemistry. The combined data set will provide high vertical scale resolution observations for validation of AURA measurements in the tropics.

## 2. Tropical Transport and the Tropical Transition Layer (TTL) Hypothesis

When considering transport of trace constituents it is useful to consider the troposphere-stratosphere system divided into regions of rapid "isentropic mixing" that are separated by semi-permeable "mixing barriers" (of which the tropopause is one), as illustrated in

91  Figure 7-6 of WMO (1999). In this framework, the tropical troposphere is viewed as
92  well-mixed, with convection rapidly transporting air to (and occasionally across) the
93  tropical tropopause. Above the tropical tropopause there is slow ascent, which is
94  primarily in response to extratropical wave driving in the stratosphere (the "stratospheric
95  pump") (Holton, 1995). In addition to these vertical motions there is isentropic mixing
96  across the subtropical tropopause, which transports lowermost stratospheric air into the
97  tropical UT, and across the stratospheric subtropical barrier (in the (isolated) "tropical
98  pipe" paradigm this mixing is exclusively out of the tropics). While this conceptual
99  picture has been extremely useful for understanding stratospheric transport and its role in
100 determining the distribution of trace constituents, research in recent years indicates that it
101 is incomplete and modifications are needed, particularly near the tropical tropopause.
102
103 Although convection has been observed to penetrate the tropical tropopause, this is not a
104 common occurrence and tropical deep convection rarely penetrates above 14 km. So
105 rather than considering the whole tropical troposphere to be well-mixed by convection, it
106 is may be better to consider convection to mix air up to around 14 km, with slow ascent
107 (driven by the stratospheric "pump") above this altitude (Highwood and Hoskins, 1998;
108 Folkins *et al.*, 1999).
109
110 Together with this slow ascent there is also mixing across the subtropical tropopause,
111 driven primarily by tropospheric weather systems (in particular, the summer monsoons)
112 (e.g. Chen, 1996; Postel and Hitchman, 1999). These systems not only mix lowermost
113 stratospheric air and tropical UT air, but they also have an effect in the lowest altitudes of
114 the overworld (where all isentrops exist solely within the stratosphere (Holton, 1995).
115 Analyses of trace gas and aerosol observations indicate there is rapid transport out of the
116 tropics in the lowest 2 km of the stratosphere (Trepte *et al.,* 1993; Rosenlof *et al.*, 1997;
117 Boering *et al.*, 1995; Mote *et al.*, 1998). More importantly for understanding the tropical
118 budgets of constituents, there is mixing of mid-latitude air into the region of tropical
119 ascent [especially below ~20 km (440K)] (*e.g.*, Avallone and Prather, 1995;
120 Minschwaner *et al.,* 1996, Volk *et al.*, 1996; Flocke *et al.*, 1999).
121
122 Taken together the above studies imply that, rather than thinking of a definite boundary
123 between the troposphere and stratosphere at the tropical tropopause, it is more
124 appropriate to think of a tropical transition layer ("TTL") that extends from around 2 km
125 below the tropical tropopause (i.e. top of typical deep convection) to around 2 km above
126 the tropopause (Highwood and Hoskins, 1998; Folkins *et al.,* 1999). The TTL is
127 illustrated schematically in *Figure 2.1*. Within the TTL there is slow ascent as well as
128 quasi-horizontal mixing with the extratropical stratosphere (both in the middle and
129 overworld), and some (probably small) degree of convective overshooting and mixing.
130 Such a transition layer, with slow ascent and some mixing of stratospheric air into the
131 tropical UT is consistent with observations of a "standing reserve" of stratospheric air in
132 the tropical UT (Tuck *et al.*, 1997) and with observed vertical profiles of total particle
133 mixing ratios in the tropics (Brock *et al.*, 1995).
134
135 The above picture is based on a zonally averaged view of the circulation and constituent
136 distributions, but there are notable zonal asymmetries within the TTL (*Figures 2.2* and

137    *2.3*). The most obvious spatial variations are due to the occurrence of regions of strong
138    convection, which result in large spatial, and seasonal, variations in the ascent into (and
139    temperature within) the TTL.  However, there are also important variations in horizontal
140    flow and lateral transport, which are linked to above variations in vertical motion and
141    regional tropospheric systems. Of particular note are the summer subtropical monsoons,
142    and the equatorial westerly ducts that exist during northern winter.

143

144    The summer subtropical monsoons have a strong impact on the transport and composition
145    of the TTL. Several studies have shown that upper-level anticyclones associated with the
146    monsoons are very important for transport across the subtropical tropopause (Chen, 1996;
147    Postel and Hitchman, 1999; Dethof *et al.*, 1999). In particular, there is more transport
148    across the tropopause during summer, with the transport (wave breaking) occurring
149    predominantly downstream of the monsoons. The monsoon anticyclones extend into the
150    stratosphere and not only influence STE but also tropical-extratropical exchange in lower
151    stratosphere (Trepte *et al.,* 1993; Horinouchi *et al.*, 2000), i.e., the monsoons can have an
152    influence throughout the depth of the TTL. As discussed below, the monsoons may have
153    a particularly large impact on both UT and LS water vapor.

154

155    Another important regional circulation feature is the occurrence of equatorial westerlies
156    over the Pacific and Atlantic Oceans, at all altitudes within the TTL, during northern
157    winter (Tomas and Webster, 1994), as shown in *Figure 2.2*. [During northern summer
158    there are easterlies at all longitudes (*Figure 2.3*)]. These "westerly ducts" are preferred
159    regions for tropical-extratropical interactions (*e.g.*, Tomas and Webster, 1994), and in
160    particular appear to be preferred regions for mixing into and within the TTL (Waugh and
161    Polvani, 2000; Horinouchi *et al.*, 2000). Furthermore, west of these ducts there is an
162    anticyclonic circulation between the equatorial easterlies and subtropical westerly jets,
163    which transports air into and out of the moist regions in UT and the tropopause cold pool
164    over Indonesia and Western Pacific (and hence will impact both the UT subtropical
165    moisture and stratospheric water vapor, see below).

166

167    In addition to the seasonal variations discussed above there are also interannual
168    variations. There is likely to be a strong influence of both the QBO and ENSO on the
169    TTL (through location of tropical convection as well strength of monsoons and the
170    equatorial westerly ducts). Furthermore, changes in the strength of the stratospheric
171    circulation (such as those inferred from trends in upper stratospheric $CH_4$, e.g., Nedoluha
172    *et al.*, 1998; Randel *et al.*, 1999) are also likely to influence the transport and composition
173    within the TTL.

174

175    In summary the **TTL hypothesis for transport within tropical UT and LS** is:

176

177    1.  There is a layer between approximately 14 and 20 km in which there is slow ascent
178        (driven by extratropical wave driving in the stratosphere) and quasi-horizontal mixing
179        with the extratropics.

180

181  2. The lateral mixing into/out of/within this layer is limited to certain regions and
182     seasons: downstream of the summer monsoons, and within the northern winter
183     westerly ducts (over the Pacific and Atlantic oceans).
184
185  If this hypothesis is correct,
186
187     • The composition in the upper 2 to 3 km of the tropical troposphere will not be
188        uniform, and will be a mixture of air from different locations (the surface, the
189        stratosphere, and the free troposphere).
190
191     • There will be a significant signature of middle latitude air in the first 2 km above
192        the tropical tropopause.
193
194     • There will be seasonal and longitudinal variations in trace constituents within the
195        tropical UT and LS that are strongly coupled to seasonally varying, regional
196        tropospheric systems.
197
198  The measurement strategy proposed in the following sections to address issues regarding
199  stratospheric and upper tropospheric water vapor and the photochemistry in the UT/LS
200  will enable the above issues to be tested.  Measurements of tracers with a range of
201  lifetimes will provide (via both profiles and tracer-tracer relations) information on
202  dominant transport pathways to the different regions (i.e. the fractions of air in a given
203  region that has been mixed rapidly from the surface or lateral mixed into the region).
204

## 3. Stratospheric Water Vapor

205
206
207  *3.1. Introduction*
208
209  A significant fraction of the observed ozone depletion during the past 2 decades in polar
210  regions and at mid-latitudes is clearly associated with the rise in chlorine and bromine
211  due to anthropogenic release of CFCs, halons, and other halocarbons (WMO, 1999:
212  Chapter 12). Industrial production of these pollutants has essentially been banned by the
213  Montreal Protocol and its amendments.  If "all else remains equal", the ozone layer will
214  recover as chlorine and bromine levels fall.  As discussed below, increases in
215  stratospheric $H_2O$ may complicate the expected recovery of the $O_3$ layer.
216
217  Observations of $H_2O$ by ground based microwave instruments (Peter, 1998), by balloon-
218  borne frost point hygrometers (Oltmans and Hoffman, 1995), and by the Halogen
219  Occultation Experiment (HALOE) satellite instrument (Evans *et al.*, 1998) all show
220  evidence for a rise in the mixing ratio of $H_2O$ during the 1990s.  The time series for $H_2O$
221  exhibits non-linear variations due to numerous forcings and the computed linear trend for
222  $H_2O$ is sensitive to the time period of the data record, the range of altitudes and latitudes
223  under consideration, and to the statistical technique used to extract the linear component
224  from the signal.  The HALOE observations reveal a rise in $H_2O$ between 1992 and 1996
225  of 55 to 150 ppbv/yr, as shown in *Figure 3.1*.  This change is much larger than can be

226 attributed to the slow rise in tropospheric $CH_4$ (WMO, 1999: Chapter 6).  The rate of
227 increase of stratospheric $H_2O$ observed by HALOE has slowed subsequent to 1996, but
228 HALOE still records a substantial increase between 1992 and 1999 (*Figure 3.2*) (Smith *et*
229 *al.*, 2000).

231  The record for $H_2O$ is not clear, however. Observations from SAGE II show a slight
232 decrease in stratospheric $H_2O$ over the 1990s (Rosenlof *et al.*, 2000).  Observations of
233 $H_2O+2CH_4$ from instruments aboard the ER-2 also indicate the lack of a significant trend
234 between 1993 and 1997 (*Figure 3.3*) (Hurst *et al.*, 1999).  Most interestingly, the HALOE
235 trend for $H_2O$ in the lower stratosphere between 1992 and 1999 shown in *Figure 3.2* is
236 rather small, perhaps consistent with the ER-2 measurements. Perhaps the larger trend for
237 $H_2O$ at higher altitudes reported by HALOE reflects conditions for older air than was
238 sampled by the ER-2.

240 An increase in the abundance of stratospheric $H_2O$ will, all else being equal, lead to a
241 decrease in the concentration of $O_3$.  At mid-latitudes, $H_2O$ supplies $HO_x$ radicals that
242 directly catalyze removal of $O_3$ (e.g., McElroy *et al.*, 1992; Wennberg *et al.*, 1994).
243 Higher levels of $H_2O$ exacerbate $O_3$ loss by promoting more efficient chlorine activation
244 by sulfate aerosols in the lower stratosphere and by promoting formation of polar
245 stratospheric clouds at warmer temperatures in both the Antarctic and Arctic circumpolar
246 vortices (e.g., Kirk-Davidoff *et al.*, 1999).  If a 100 ppbv/yr trend in $H_2O$ (the trend
247 observed between 1992 and 1996) were to persist until 2010, the expected recovery of
248 stratospheric $O_3$ due to the ~15% decline in chlorine loading would be negated by
249 increased $O_3$ loss from $HO_x$ radicals (Jucks and Salawitch, 2000).

251 In general terms the aridity of the stratosphere is clearly due to the cold tropical
252 tropopause (Brewer, 1949).  However, the precise physical mechanism(s) responsible for
253 the aridity of the lower stratosphere are unclear.  Various "Hypotheses of Stratospheric
254 Aridity" are described in the following section, and proposed observational tests of these
255 hypotheses are given in Section 3.5.

257 One of the motivations of the proposed field campaign is that the cause of the recent rise
258 in stratospheric $H_2O$ is not understood.  A simple explanation for this rise is a warming of
259 the coldest regions of the tropical tropopause since 1992 (e.g., Evans *et al.*, 1998).
260 However, an analysis of a time series (from 1979 to present) of temperature near the
261 tropical tropopause, shown in *Figure 3.4*, reveals a general cooling trend from 1992 to
262 1998 throughout the tropics and a slight cooling trend in the Western Pacific (Simmons *et*
263 *al.* 1999).  The tropical-mean trend is most pronounced between 1992 and 1996, when
264 ground-based and HALOE observations suggest stratospheric $H_2O$ increased quite
265 rapidly.  In section 3.3, we describe several "Hypotheses of Stratospheric $H_2O$ Change"
266 that are decoupled from minimum tropopause temperature.  Since future changes in
267 stratospheric $O_3$ cannot be forecast reliably until we have a better understanding of the
268 processes that regulate stratospheric $H_2O$, it is essential that observational tests of these
269 hypotheses be designed and carried out.

272 *3.2 Hypotheses of Stratospheric Aridity*

274 The mixing ratio of stratospheric $H_2O$ is controlled by processes in the tropical upper
275 troposphere and lowermost stratosphere. In general terms the aridity of the stratosphere
276 is clearly due to the cold tropical tropopause. However, the precise physical mech-
277 anism(s) responsible for the aridity of the lower stratosphere are unclear.

279 Various hypotheses have been proposed to account for details of stratospheric aridity.
280 There is no commonly accepted set of definitions or nomenclature for these hypotheses.
281 The definitions and names given below represent an attempt to distill the essence of
282 hundreds of papers that have been written on this topic during the past 50 years and are
283 used to motivate the observational plans described in the following sections. It is beyond
284 the scope of this document to delve into great detail regarding the numerous
285 observational details in support of, or refuting, the various hypotheses. Summaries of
286 these observational details are given by Holton *et al.* (1995), Mote *et al.* (1996), Moyer *et*
287 *al.* (1996), Keith (2000), and Sherwood and Dessler (2000). Suffice to say there is no
288 commonly accepted theory for how the aridity of the stratosphere is maintained.

290 Class I: One dimensional models of stratospheric $H_2O$. Each of these hypotheses treats
291 the entry of air into the stratosphere, and the desiccation of stratospheric air, as events
292 that happen essentially simultaneously during vertical ascent in the tropics.

294     1) **Cold Trap** - slow, large-scale upwelling through the cold tropopause dehydrates
295        ascending air (Brewer, 1949)

297        1a) *Fountain* - air enters the stratosphere only in the Western Pacific, the coldest
298            regions of the tropopause (Newell and Gould-Stewart, 1981)

300     2) **Convective Overshoot** - Convective turrets ascend rapidly from near the ground
301        and penetrate into the stratosphere, where they induce rapid dehydration due to
302        precipitation of ice (Danielsen, 1982)

304        2a) *Ice Lofting* - air entering the stratosphere contains significant amounts of ice,
305            some of which precipitates and some of which evaporates (Johnston and
306            Solomon, 1979).

308        2b) *Supersaturation* - air entering the stratosphere is highly supersaturated (*e.g.*,
309            150%) with respect to cloud top temperature (Moyer *et al.*, 1996)

311 Class II: Three dimensional models of stratospheric $H_2O$. These hypotheses treat the
312 entry of air into the stratosphere, and the desiccation of stratospheric air, as events that
313 happen at different times and in different places. A feature of these hypotheses is
314 recognition that processes (such as precipitation and mixing) within the TTL may play an
315 important role in determining stratospheric $H_2O$.

3) **TTL Cold Trap** - air enters the TTL to the west of the region of coldest temperatures in an area of forced upwelling and parcels are advected to the region of coldest TTL temperatures by an anti-cyclonic flow. As the air approaches the region of coldest temperature, it sinks, cools, and desiccates over relatively long time scales (several days). Downstream of the TTL cold trap, the air is heated diabaticaly and rises, eventually mixing into the stratosphere (J. R. Holton, private communication, 2000).

4) **TTL Overshoot** - convective turrets penetrate the TTL. The more energetic turrets penetrate to higher levels and become rapidly (time scales of less than a day) dehydrated due to sedimentation of ice. The resulting humidity of the TTL is caused by mixtures of air parcels that detrain at different potential temperatures. Slow, zonally uniform ascent carries desiccated TTL air into the stratosphere (Sherwood and Dessler, 2000).

5) **Stratospheric Dehydration** - Dehydration in the lowermost stratosphere is induced by meso or synoptic-scale wave activity that lifts and cools air masses in the lower stratosphere (Fritsch and Brown, 1982; Teitelbaum *et al.*, 2000).

The **Cold Trap** theory was proposed to explain the overall aridity of the stratosphere (Brewer, 1949). It was later realized that the water vapor mixing ratio of air entering the stratosphere (currently considered to be about 3.8 to 4.0 ppmv: Abbas *et al.*, 1996; Dessler, 1998) is drier than can be accounted for by the mean temperature of the tropical tropopause, prompting the proposed *Fountain* corollary to the original theory (Newell and Gould-Stewart, 1981). In both cases, stratospheric aridity is maintained by exposure of air to the coldest regions of the tropical tropopause as these parcels first enter the stratosphere.

The **Convective Overshoot** theory was based on the recognition that some cumulonimbus clouds penetrate the tropopause and can irreversibly mix with stratospheric air (Danielsen, 1982). Data from the NASA STEP (Stratosphere-Troposphere Exchange Program) mission provided evidence for a single occurrence of such an event (Danielsen, 1993). The *Ice Lofting* and *Supersaturation* corollaries account for the observation that the water vapor mixing ratio of air entering the stratosphere is typically wetter than can be accounted for by the exceedingly cold temperatures at the top of convective turrets.

Both the **TTL Cold Trap** (J. R. Holton, private communication, 2000) and the **TTL Overshoot** (Sherwood and Dessler, 2000) hypotheses were motivated in part by recent meteorological analyses that indicate the cold tropical Western Pacific region is an area of net subsidence near the tropopause (Gettelman *et al.*, 2000; Sherwood, 2000). This is the same geographic area for which Newell and Gould-Stewart (1981) hypothesized upwelling as a key part of the *Fountain* corollary. *One of the primary goals of our proposed mission is to test this recent suggestion of mean downward motion in the Western Pacific tropopause region by measuring mixing ratio profiles of short lived tracers during N.H. winter* (the time of coldest temperature).

363

364 The energetic turrets of the **TTL Overshoot** penetrate the bottom of the TTL, but do not
365 reach the true stratosphere (the overworld). Dehydration occurs rapidly by sedimentation
366 of ice as air ascends vertically: rapid dehydration is essential because the overshooting
367 turrets remain exceedingly cold for only brief periods of time. In contrast, the **TTL Cold**
368 **Trap** theory hypothesizes that dehydration occurs much more slowly (e.g., over a period
369 of several days) as air is advected horizontally (quasi-isentropically) through a standing
370 cold pool. Consequently, stratospheric $H_2O$ is most likely sensitive to cloud
371 microphysics according to the **TTL Overshoot** theory and to details of transport
372 according to the **TTL Cold Trap** scenario. For both hypotheses, dehydration of air
373 occurs prior to its arrival into the overworld.

374

375 Finally, the **Stratospheric Dehydration** theory was proposed in part to account for the
376 observation that the minimum mixing ratio of stratospheric water often occurs 2 to 3 km
377 above the tropopause in both the tropics and mid-latitudes (e.g., Kley *et al.*, 1979). Mote
378 *et al.* (1996) have shown, however, that this feature can also be accounted for by the
379 seasonal cycle of tropopause temperature being "recorded" onto the vertical profile of
380 $H_2O$ due to slow uplift in the tropical lowermost stratosphere.

381

382 Before offering a list of "Hypotheses of Stratospheric $H_2O$ Change", we digress to
383 consider how stratospheric $H_2O$ might respond to various forcings for each of the
384 "Hypotheses of Stratospheric Aridity" defined above. This digression is meant to
385 motivate the usefulness of pursuing tests of the various "Hypotheses of Stratospheric
386 Aridity" as a first step for understanding the cause of recent changes to stratospheric
387 $H_2O$.

388

389 For the various hypotheses, we expect:

390

391    **Cold Trap** - stratospheric $H_2O$ will be most sensitive to tropopause temperature

392

393    **Convective Overshoot** - stratospheric $H_2O$ will be most sensitive to surface
394        temperature and humidity (which determine turret height), the amount of ice
395        lofted across the tropopause, perhaps ice physics, and only indirectly to
396        tropopause temperature

397

398    **TTL Cold Trap** - stratospheric $H_2O$ will be most sensitive to temperature in the
399        TTL, circulation patterns within the TTL, and transport of air into and out of the
400        TTL.

401

402    **TTL Overshoot** - stratospheric $H_2O$ will be most sensitive to surface temperature and
403        humidity (which determine turret height), perhaps ice physics, circulation patterns
404        within the TTL, and transport of air into and out of the TTL, and only indirectly to
405        tropopause temperature.

406

407    **Stratospheric Dehydration** - stratospheric $H_2O$ will be most sensitive to lower
408        stratospheric temperature and transport process in the lower stratosphere.

409

*3.3 Hypotheses of Stratospheric H₂O Change*

The various "Hypotheses of Stratospheric Aridity" described above have different consequences for how stratospheric $H_2O$ is likely to respond to changing climate. Understanding precisely how stratospheric aridity is maintained for a brief period in time is not necessarily sufficient for understanding how stratospheric $H_2O$ may change over multi-year time periods. The proposal to conduct this mission over an extended period of time is motivated by the need to gain insight into the seasonal and interannual variations in the transport of $H_2O$ within the TTL.

The following hypotheses have been proposed to explain the recent rise in stratospheric $H_2O$. For convenience of distinguishing these hypotheses from the above mentioned "Hypotheses of Stratospheric Aridity", the numbering sequence starts with "6":

6) **Rising Cold Trap Temperature** - Stratospheric $H_2O$ is responding directly to a warming of the tropopause region (Evans *et al.*, 1998).

7) **Increasing Asian Monsoon** - An increase in the extremity of the Asian monsoon over Indonesia leads to a moister upper troposphere, resulting in greater abundance of $H_2O$ within the TTL for air parcels that do not pass directly through a cold trap (Evans *et al.*, 1998; Smith *et al.*, 2000).

8) **Changing Ice Physics** - Convective turrets become less efficiently dehydrated over time due to subtle changes in the microphysics of ice (an adaptation of Sherwood and Dessler, 2000)

9) **Reduced Brewer-Dobson Circulation** - Stratospheric wave activity in the NH is reduced, resulting in increased fraction of air entering the TTL during NH summer (the warm tropopause period) (this white paper).

The radiosonde analyses presented by Simmons *et al.* (1999) appear to contradict the **Rising Cold Trap Temperature** theory. Smith *et al.* (2000) present analyses of HALOE data that indicate a considerable moistening of the tropical upper troposphere in September-October-November between 1992 and 1999 that support the **Increasing Asian Monsoon** theory. However, it is an open question whether or not this moistening of the upper troposphere has a direct effect on stratospheric $H_2O$. It is unclear why or how the microphysics of ice would change from year to year, and Sherwood and Dessler (2000) mention the sensitivity of stratospheric $H_2O$ to ice physics only "in passing". The extreme year-to-year variability in the extent of denitrification of the Arctic polar vortex suggests ice physics *could* play a role in year-to-year variations of stratospheric $H_2O$.

Finally, the **Reduced Brewer-Dobson Circulation** theory is offered as another mechanism for increasing stratospheric $H_2O$. It is conceivable that rising concentrations of greenhouse gases may have induced a reduction in the "extratropical pump" (Holton *et al.*, 1995) altering the Brewer circulation for essentially the same reasons as discussed by

455 Shindell *et al.* (1998) for the polar vortex extension of this circulation. According to the
456 framework of Yulaeva *et al*. (1994), a slow down of the Brewer-Dobson circulation
457 should be associated with tropopause cooling. However, it is unclear whether tropopause
458 temperature is governed more strongly by local thermodynamics (e.g., Kirk-Davidoff *et*
459 *al.*, 1999) or by large-scale transport. Stratospheric $H_2O$ is likely the result of an
460 admixture of processes that occur mainly during NH winter but also somewhat during
461 summer. A slight increase in the relative contribution of summertime processes (when
462 temperature in the tropical upper troposphere is warmer) to resulting overworld air,
463 perhaps due to a change in the *seasonal variation* of the Brewer-Dobson circulation,
464 could contribute to the recent rise in stratospheric $H_2O$.
465
466
467 *3.4 The Importance of Isotopic Measurements for testing the hypotheses*
468
469 Measurements of the isotopic composition of water vapor comprise a key aspect of our
470 proposed strategy for testing the various hypotheses of stratospheric aridity described
471 above. Here we describe how measurements of water vapor isotopic composition will be
472 used. The material in this section is based largely on the work of Moyer *et al.* (1996) and
473 Webster *et al.* (2000).
474
475 The isotopic composition of $H_2O$ records the condensation and evaporation history
476 experienced by air parcels. In thermodynamic equilibrium, condensates of $H_2O$ are
477 isotopically enriched (e.g., heavier) relative to gas phase $H_2O$. Consequently, as water
478 condenses and precipitates from an ascending air parcel, the residual vapor becomes
479 progressively lighter. The abundance of HDO relative to $H_2O$ (termed $\delta D$) and of $H_2^{18}O$
480 relative to $H_2O$ (termed $\delta^{18}O$) can be calculated for an ascending air parcel using
481 fractionation factors between ice and vapor based on laboratory measurements (Merlivat
482 and Nief, 1967; Majoube, 1970), assuming an ambient temperature profile from
483 observation and that $\delta D$ and $\delta^{18}O$ at cloud base equals that of the sea surface (e.g., Moyer
484 *et al*., 1996; Keith, 2000; Johnson *et al.*, 2000).
485
486 Moyer *et al.* (1996) showed that the expected value of $\delta D$ at 100 mbar in the tropics is
487 about -900 per mil (*Figure 3.5*). This calculation assumed thermodynamic equilibrium,
488 desiccation to observed levels of stratospheric aridity, and no transport of condensed
489 material across the tropopause. However, ATMOS measured $\delta D$ of -670 per mil in the
490 tropical lower stratosphere (Moyer *et al.*, 1996). Johnson *et al.* (1995), using balloon-
491 borne thermal emission infrared spectrometer measurements obtained at mid-latitudes,
492 estimated a similar value of $\delta D$ for air entering the stratosphere. Minor contributions to
493 HDO from oxidation of $CH_3D$ have been accounted for in both estimates of $\delta D$.
494
495 The observations of $\delta D$ appear to be inconsistent with the **Cold Trap** hypothesis of
496 stratospheric aridity (Moyer *et al*., 1996). Moyer *et al*. (1996) suggested two possible
497 explanations of the measurements. First, air parcels in the tropical lower stratosphere
498 may receive contributions to their $H_2O$ budget from isotopically enriched ice lofted
499 across the tropopause that later evaporates (the **Convective Overshoot - Ice Lofting**
500 hypothesis discussed above). The second possibility is that the actual isotopic

fractionation that occurs in the atmosphere may be weaker than the equilibrium value, which would occur during ice deposition for highly supersaturated conditions (e.g., saturation ratios of 150 to 200%) (the **Convective Overshoot - Supersaturation** hypothesis discussed above).

Keith (2000) extended the work of Moyer *et al.* (1996) and noted two key points. First, the kinetic fractionation factors used by Moyer *et al.* (1996) are based on laboratory measurements obtained for temperatures of 240 K and above. Results are extrapolated to colder temperatures. Keith (2000) states "it seems unlikely that errors in the isotopic physics are of sufficient magnitude to resolve the HDO puzzle". Nonetheless, laboratory measurements must be obtained at colder temperature to better interpret the isotopic measurements proposed here. The second point raised by Keith (2000) is the illustration that an estimate of $\delta^{18}O$ for air entering the stratosphere from the mid-latitude balloon measurements of Johnson *et al.* (2000), when combined with the $\delta D$ measurements of Moyer et al. (1996) and Johnson *et al.* (1995), appear to rule out the **Convective Overshoot - Supersaturation** hypothesis of stratospheric aridity. Thus, the **Convective Overshoot - Ice Lofting** theory is the only Class I hypothesis that is consistent with the limited isotopic measurements of $H_2O$.

There are many caveats to the conclusions noted above. The ATMOS measurements of HDO in the tropics are sparse and do not extend to the upper troposphere. *Continuity of profiles of $\delta D$ across the tropopause is essential*. The FIRS estimates of $\delta D$ and $\delta^{18}O$ of air entering the stratosphere are based on measurements obtained only at mid-latitudes. *Measurements of $H_2^{18}O$ in the tropics are needed*. Current theoretical models of $\delta D$ and $\delta^{18}O$ only treat stratospheric dehydration in one dimension (e.g., the Class I hypotheses described above). *The consequences of condensation, lofting, and mixing within the hypothesized TTL must be incorporated into a more sophisticated theoretical treatment of $\delta D$ and $\delta^{18}O$*. Although the existing observations appear to support the **Convective Overshoot - Ice Lofting** hypothesis, more atmospheric measurements of $\delta D$ and $\delta^{18}O$ are needed. For example, the current remote observations of $\delta D$ and $\delta^{18}O$ have insufficient vertical resolution to address the TTL "Class II" hypotheses of stratospheric aridity.

The discussion so far has focused only on differences in vertical profiles of $\delta D$ and $\delta^{18}O$ predicted by the various "Class I" hypotheses of stratospheric aridity. However, the three "Class II" (e.g., three-dimensional) hypotheses will lead to differences in spatial patterns of $\delta D$ and $\delta^{18}O$ as well. The **TTL Cold Trap** hypothesis should result in considerable horizontal gradients in $\delta D$ and $\delta^{18}O$ as air flows through the cold trap, and relatively gentle vertical gradients across the lower and upper boundary of the TTL. The **TTL Overshoot** hypothesis would be consistent with strong vertical gradients in $\delta D$ and $\delta^{18}O$ at the lower boundary of the TTL, gentle horizontal gradients within the TTL, and gentle vertical gradients near the top of the TTL. The **Stratospheric Dehydration** hypothesis will predict gentle horizontal and vertical gradients in $\delta D$ and $\delta^{18}O$ until truly stratospheric air is encountered.

Perhaps the strongest use of measurements of $\delta D$ and $\delta^{18}O$, which has not yet been explored in the literature, will be the *potential to quantify the effects on $H_2O$ of both*

547     *thermodynamics and mixing* for air parcels with different temperature histories within the
548     hypothesized TTL. Correlation plots of $H_2O$ vs $\delta D$ and $H_2O$ vs $\delta^{18}O$ will be highly non-
549     linear for individual ascending air parcels. Therefore, admixtures of slowly ascending air
550     masses that detrain at different potential temperatures within the TTL (*e.g.*, Sherwood
551     and Dessler, 2000) may best be identified by the appearance of "mixing lines" (e.g.,
552     Waugh *et al.*, 1997) in correlation plots of $H_2O$ vs $\delta D$ and $H_2O$ vs $\delta^{18}O$. Since $\delta D$ and
553     $\delta^{18}O$ essentially preserve a memory of thermodynamic changes in ambient $H_2O$ that is
554     strongly dependent on the coldest recent temperature, correlation plots of $H_2O$ vs its
555     isotopes may ultimately prove to be the best way to quantify the effects on $H_2O$ of both
556     thermodynamics and mixing.
557
558     *3.5 Testing the Hypotheses*
559
560     The observations of $H_2O$ and temperature that will be obtained by HIRDLS, MLS and
561     TES in the tropical tropopause region will provide important information on the
562     longitudinal variability of conditions in the upper troposphere and lower stratosphere.
563     The cirrus ice measurement from MLS may provide important constraints on the details
564     of whether significant amounts of precipitable water vapor are lofted across the
565     tropopause. Space-borne observations of $H_2O$ will also be obtained in the future by
566     SAGE III. To our knowledge, TES is the only upcoming space-based instrument that
567     will attempt to measure HDO. Other satellites payloads may have some capability (e.g.
568     SCIAMACHY) but have not yet focused their efforts on isotope retrievals.
569
570     Our mission has four primary goals for testing hypotheses of both stratospheric aridity
571     and hypotheses of long-term changes in stratospheric $H_2O$:
572
573     A. Testing the accuracy of the AURA (and other satellite) measurements of $H_2O$ and
574     temperature in the tropical tropopause region;
575
576     B. Determination of the vertical and horizontal structures of fields of $H_2O$, HDO, and
577     temperature in the tropics and whether these structures are suitably resolved by the
578     satellite instruments;
579
580     C. Testing the accuracy of the MLS cirrus ice measurement;
581
582     D. Measurement at high vertical resolution of the isotopic composition of $H_2O$ and
583     certain tracers of vertical motion that have a wide range of lifetimes (e.g., $SF_6$, $CO_2$, CO,
584     $CH_3I$, radon) to complement the satellite measurements of $H_2O$, HDO, and other longer-
585     lived tracers.
586
587     ***Note: we have not addressed at all in this draft white paper the issue of the local***
588     ***radiative balance in the tropics. Clearly this is an important issue. We hope to receive***
589     ***feedback from the community regarding the incorporation of measurements of the***
590     ***radiative field into the framework of this proposed mission.***
591

592 The importance of accurate measurements of $H_2O$ for testing the various hypotheses of
593 stratospheric aridity is obvious. It is essential for the achievement of goal A that the
594 correlative measurements be obtained in the tropics. A variety of correlative
595 measurement techniques will likely be required. Lidar measurements of $H_2O$ and
596 microwave temperature profiler measurements of temperature will provide greatest
597 flexibility for obtaining close coincidence in space and time. Airborne in situ
598 measurements of temperature will provide the greatest possible accuracy. Airborne in
599 situ measurements of $H_2O$ using existing instruments will provide a capability for
600 continuity between AURA and UARS measurements of $H_2O$, since there are numerous
601 coincidences between HALOE and MLS measurements of $H_2O$ and observations from
602 previous in situ campaigns. Balloon measurements of $H_2O$ and temperature in the tropics
603 are highly desirable because satellite measurements of $H_2O$ above the cruise altitude of
604 either the ER-2 and WB-57 are of interest (e.g., for assessing the seasonal cycle of $H_2O$)
605 and it is not clear the lidar measurements will be sufficiently accurate at these altitudes to
606 be the sole source of validation.

608 For the achievement of goal A), it will be highly desirable to continue to explore the
609 cause of lingering differences between measurements of $H_2O$ by various in situ
610 instruments (differences sometimes as high as 15%). Laboratory intercomparisons of the
611 various in situ instruments should be planned as part of this mission.

613 Goal B: Testing theories of stratospheric aridity will require accurate knowledge of the
614 vertical and longitudinal variability of $H_2O$ and HDO in the tropics. Determination of
615 these vertical and horizontal structures, and whether these structures are suitably resolved
616 by the space-based instruments, necessitates obtaining correlative observations over both
617 the Western Pacific (cold tropopause) and the Eastern Pacific (warmer tropopause). It is
618 critical that we assess how well the various satellite instruments can resolve the shape of
619 the vertical profile of $H_2O$ in the regions where we expect the largest vertical gradients
620 and the minimum abundance of $H_2O$ to occur (e.g., the Western Pacific). Understanding
621 thermodynamics and transport within the TTL will require testing the ability of the
622 satellite instruments to distinguish subtle differences in composition between the
623 poleward subtropical jet and the equatorial easterly region of the tropical anti-cyclonic
624 flow in the Western Pacific. Consequently, we propose a series of airplane flights from
625 Guam (13.4° N, 144.4° E) and Hawaii (21.3° N, 157.9° W) to provide access to all of the
626 regions of interest.

628 It is important that we assess how well the various satellite instruments can resolve the
629 shape of the vertical profile of $H_2O$ in the regions where the minimum abundance of $H_2O$
630 is expected to occur (e.g., the Western Pacific). The importance of this validation activity
631 is illustrated by a comparison of vertical profiles of $H_2O$ obtained by HALOE and an in
632 situ hygrometer in the tropics during October 1994. Figure 1 of Mote *et al.* (1996)
633 revealed differences of nearly 0.7 ppmv in the minimum mixing ratio of $H_2O$ (the
634 HALOE measurement of $H_2O$ exceeded the in situ observation) that appears attributable
635 to the finer altitude resolution of the in situ measurement. This difference compromises
636 the ability of HALOE to accurately identify the $H_2O$ mixing ratio content of air that
637 enters the stratosphere. Since one of the strongest tests of the various hypotheses of

638 stratospheric aridity is the relation between minimum tropopause temperature (or
639 minimum TTL temperature) and the $H_2O$ mixing ratio of newly minted stratospheric air,
640 it is essential that our validation activitiies be focused on assessing the reliability of the
641 space-borne measurements of minimum $H_2O$ and temperature.

643 It is important also to obtain measurements of $H_2O$ and temperature in the NH summer as
644 well as during the NH winter (coldest tropopause) because the mechanisms for cross
645 tropopause transport may be distinctly different. The humidity of the stratosphere
646 reflects the mixing ratio of water entering the stratosphere weighted by the mass flux
647 through the tropical tropopause. Although ascent is significantly slower in NH summer
648 than during NH winter (*e.g.*, Rosenlof *et al.*, 1997), the much higher water mixing ratio at
649 the tropopause yields a mass flux of water that significantly humidifies the stratosphere.
650 This is seen in the seasonal cycle of water in the region of tropical ascent (Mote *et al.*,
651 1996). Stratospheric humidity may thus be influenced by interannual changes in the
652 seasonal variation of mass flux through the tropopause, independent of any change in the
653 mixing ratio of water at the tropical tropopause.

655 The importance of isotopic measurements of $H_2O$ for testing hypotheses of stratospheric
656 $H_2O$ is described in the previous section. The only $H_2O$ isotopic measurement from EOS
657 Aura will be profiles of HDO provided by TES. For validation of this measurement, the
658 existing balloon borne remote measurement capability is furthest developed. Profiles of
659 HDO have been published by the FIRS group (Johnson *et al.*, 2000). The MkIV
660 instrument can measure profiles of HDO with better precision than ATMOS owing to the
661 longer integration time from a balloon platform (G.C. Toon, private communication,
662 2000). While balloon-borne observations of HDO should play an important role in
663 testing the accuracy and vertical resolution of the TES observation, these measurements
664 can not be obtained in the Western Pacific region unless a water-landing capability for
665 the gondola is developed.

667 Various options are available for air-borne measurements of $H_2O$ isotopes. A tunable
668 diode laser instrument for measurement of HDO and $H_2^{18}O$ from the WB-57 is currently
669 under development at JPL (Webster *et al.*, 2000). Measurements of HDO and $H_2^{18}O$ can
670 also be obtained using an in situ mass spectrometer but we are unaware of any such
671 instrument development for either the ER-2 or WB-57 platforms. Owing to the critical
672 importance of water isotopes for testing the various hypotheses, we suggest that both
673 balloon-borne remote and in situ sampling of $H_2O$ isotopes in the tropics be conducted
674 for this mission. The airborne observations will be especially important for defining
675 vertical and horizontal structures in the Western Pacific that will be an important new
676 tool for testing the various hypotheses of stratospheric aridity.

678 Goal C. The MLS measurement of the ice content of cirrus is a potentially powerful
679 observation for testing the importance of "ice lofting" within both the Class I and Class II
680 hypotheses for stratospheric aridity described above. The primary concern, however, is
681 whether the satellite measurement will provide sufficient vertical resolution. At present,
682 we do not have a means for testing this space-borne observation from either the ER-2 or
683 WB-57 platforms. Such a test would require the development of a new instrument that

684 could distinguish gas phase from condensed phase $H_2O$. The desired correlative
685 measurement could be achieved by development of an inlet for one of the existing in situ
686 $H_2O$ instruments that could, at various times, either include or exclude particulates. Since
687 the existing isotopic measurements of $H_2O$ described above appear to suggest that ice
688 lofting is truly important for supplying $H_2O$ to the stratosphere, the *development of a new*
689 *in situ capability for measuring condensed phase water should be given high priority*
690 both for validation of the MLS measurement of cirrus ice and for fully testing the various
691 hypotheses of stratospheric aridity.
692
693 Goal D. The proposed measurements are designed to complement the space-borne
694 observations for further testing of the hypotheses of both stratospheric aridity and
695 hypotheses of long-term changes in stratospheric $H_2O$. Clearly the satellite measurements
696 of $H_2O$, temperature, long-lived tracers of atmospheric transport will be critical for
697 resolving longitudinal gradients throughout the tropics, variations in stratospheric
698 humidity with respect to both altitude and latitude, as well as possible long-term changes
699 in $H_2O$ and temperature. Our strategy is to augment the space-borne observations with
700 high vertical and horizontal resolution of a host of other species in the specific region
701 where it has been hypothesized that stratospheric aridity is controlled.
702
703 The usefulness of the proposed isotopic measurements for testing the various hypotheses
704 of stratospheric aridity is discussed in Section 3.4.
705
706 Measurement of a suite of gases with short photochemical lifetimes (e.g., $CH_3I$, acetone,
707 Ra, $^{210}Pb$, and/or $^{222}Rn$) will provide a strict test of whether air is sinking over the cold
708 Western Pacific, as has been proposed by Gettelman *et al.* (2000) and Sherwood (2000).
709 Vertical profiles of CO provide quantitative measurements of ascent rates on longer time
710 scales (Herman *et al.*, 1999). Tracer-tracer correlation plots of a suite of tracers with a
711 variety of lifetimes can be used to place additional quantitative constraints on transport in
712 the tropical TTL region (*e.g.*, Flocke *et al.*, 1999).
713
714 In situ measurements of the vertical structures of gas phase and particulate $H_2O$ at spatial
715 scales that can not possibly be achieved by satellite measurement (e.g., 100 m resolution)
716 will be critical for testing details of the various hypotheses of stratospheric aridity
717 described in Section 3.2. Testing the Class I and Class II Overshoot hypotheses may
718 prove quite difficult using only space-borne observations because of the rapid variation of
719 $H_2O$ on small spatial scales (*e.g.*, Sherwood and Dessler, 2000). The **TTL Cold Trap**
720 hypothesis predicts that lower levels of $H_2O$ will be seen downwind of the standing cold
721 pool than upwind of this region. While the satellite observations should be able to
722 resolve such structures, the proposed simultaneous measurement of the numerous short-
723 lived tracers mentioned above (not measured by satellite) will provide an estimate of the
724 "age" of air parcels in the TTL. This information will be key for unraveling the
725 relationship between details of the transport field and desiccation. Finally, measurements
726 of the size distribution of the particulates, using spectrometers based on existing
727 technology that are focused on several-micron size particles, will enable rates of
728 dehydration to be estimated. This will provide a test of another important difference

729 between the **TTL Cold Trap** hypothesis (relatively slow desiccation) and the **TTL**
730 **Overshoot** hypothesis (relatively rapid dehydration).

731

732 Finally, the candidate $H_2O$ instruments (both aircraft and balloon) for this mission have
733 obtained numerous coincident measurements of $H_2O$ with the existing MLS and HALOE
734 instruments on UARS and with SAGE II. Obtaining additional coincidences close in
735 space and time with the various EOS Aura instruments may prove crucial for "bridging
736 the gap" between Aura and UARS and between Aura and SAGE II for studies of long-
737 term changes in stratospheric $H_2O$, especially if UARS and/or SAGE II are not
738 operational at the time Aura turns on.

## 4. Photochemistry of the Tropical Lower Stratosphere and Upper Troposphere

739
740

741

*4.1 Introduction*
742

743

744 The chemical properties of tropical transition layer define the chemical boundary
745 condition for the stratosphere. Despite a decade of tremendous progress in tropospheric
746 and stratospheric chemistry, the photochemistry of this region remains poorly
747 constrained. This limits our ability to understand the fates of numerous compounds (and
748 their degradation products) transported into the near tropopause region. Such knowledge
749 is required if we are to understand the impact of climate change on the stratosphere in
750 general and ozone in particular.

751

752 Until recently, the chemical precursors of the stratospheric radicals and aerosol, with the
753 notable exception of water vapor, were thought to be compounds with long tropospheric
754 lifetimes. This greatly simplified defining the chemical boundary condition for the
755 stratosphere because globally-averaged surface measurements of these long-lived
756 compounds could be used. For example, sulfur was thought to be carried solely by OCS,
757 nitrogen by $N_2O$, and halogens by the relatively long-lived halocarbons. It has become
758 increasing clear, however, that short-lived compounds transported to the tropopause
759 region of the tropics significantly alter the chemistry of the global stratosphere. The
760 amount of OCS transported across the tropoause accounts for no more than half of the
761 sulfur aerosol present in the lower and middle stratosphere (*e.g.*, Weisenstein *et al.*,
762 1997). Bromine monoxide concentrations in the lower stratosphere appear to reflect the
763 input of very short-lived bromine containing organic and perhaps inorganic compounds
764 (*e.g.*, Ko *et al.*, 1997). The concentration of reactive nitrogen, $NO_y$, and ozone are non-
765 zero at the tropical tropopause (Strahan, 1999).

766

767 Because the upper troposphere in the tropics is arid and cold, until recently, it was
768 considered to be photochemically inactive. The production rate of OH was thought to be
769 determined by the local concentrations of ozone and water vapor. This rate was
770 calculated to be very low both because the yield of $O(^1D)$ in the photolysis of ozone was
771 thought to be small due to the temperature dependence of the quantum yield and because
772 the efficiency of OH production by $O(^1D)$ is reduced at the low humidity. Observations
773 of OH in the upper tropical troposphere have significantly altered this picture (Wennberg

774   *et al.*, 1997).  The measured abundance of OH between 10 km and the tropopause is
775   nearly twice that calculated.  It has been suggested that the higher than expected levels of
776   OH result from the transport of short lived $HO_x$ precursors into the arid upper
777   troposphere. Oxygenated hydrocarbons such as acetone (Singh *et al.*, 1995) and
778   peroxides (Jaegle *et al.*, 1997) have been suggested as possible sources.  Nevertheless, a
779   closure experiment in the tropical upper troposphere has not been accomplished.
780   Independent of the source of these free radicals, the larger concentration of $HO_x$ alters the
781   calculated lifetime of CO, $SO_2$, and other compounds and may increase the production
782   rate of ozone in the UT.
783
784   The short lifetimes of these compounds significantly impact the design of the $TC^3$
785   campaign.   Because the source strengths of these compounds are likely not steady,
786   reflecting the variability in both the chemistry and the transport from the lower
787   troposphere, the sampling strategy must be optimized to assure it is representative.
788   Satellite observations are key to understanding how representative the in situ data set is of
789   the variable dynamics in the tropics.
790
791   Finally, the tropical lower stratosphere offers a unique laboratory to study the photo-
792   chemical lifetimes of short-lived compounds.  This is one of the few regions of the
793   atmosphere where the chemical and dynamical processes are separable on seasonal
794   timescales.  Herman *et al.* (1999) and Weinstock *et al.* (2000) have shown, for example,
795   that the lifetime of CO can be determined empirically by measurement of the decrease in
796   its mixing ratio with height above the tropical tropopause. The ascent rate can be
797   accurately constrained by coincident $CO_2$ and $H_2O$ measurements and the influence of
798   mixing from the subtropics can be accounted for by observations of very long-lived
799   tracers such as $N_2O$.  Dessler *et al.* (1996) have shown that the ozone production rate can
800   also be determined in this manner. This analysis can be extended to any compound with
801   tropospheric sources and irreversible sinks in the lower stratosphere. Target molecules
802   include non-methane hydrocarbons and oxygenated hydrocarbons (such as acetone),
803   PAN, $SO_2$, $H_2S$, etc.
804
805   Clearly a suite of measurements of short-lived compounds in the tropical upper
806   troposphere and lower stratosphere will greatly improve our understanding of the
807   chemistry and dynamics of this critical region. Below we outline a set of hypotheses that
808   can be tested through this approach.
809
810   *4.2 Hypothesis* - **Sources of Stratospheric Bromine:**
811
812       *Short-lived bromine containing compounds enhance significantly the concentration of*
813       *bromine monoxide in the upper troposphere and lower stratosphere.  As a result,*
814       *ozone loss rates in the lower stratosphere are significantly faster than expected.*
815
816   It has become increasing clear that the bromine budget of the stratosphere can not be
817   closed by consideration of longer-lived bromine-containing halocarbons   alone.
818   Observations in the lower stratosphere demonstrate that, in the youngest air, bromine
819   monoxide concentrations are significantly larger than expected (*Figure 4.1*) (Wamsley *et*

820    *al.*, 1998).   This result is also seen in balloon-borne solar occultation measurements
821    (Pundt *et al.*, 1997, Pfeilsticker *et al.*, 2000) and ground-based slant column observations
822    of BrO (Friess *et al.*, 1999).   These bromine monoxide measurements can be explained
823    by assuming a stratospheric boundary condition at the tropical tropopause of 3 to 5 pptv
824    $Br_y$.  It is unclear, however, what molecule or molecules carry this additional bromine.
825    Suggestions have included very short-lived bromo-carbons such as bromoform ($CHBr_3$)
826    (Dvortsov *et al.*, 1999; Schauffler *et al.*, 1999; Sturges *et al.*, 2000) as well as the
827    possibility of direct input of $Br_y$ (BrO + HOBr + $BrONO_2$) (Ko *et al.*, 1997; Pfeilsticker
828    *et al.*, 2000).   Clearly, understanding the budget and potential trends in bromine require
829    knowledge of the mechanisms for cross tropopause transport. Bromine is an active
830    catalyst in the lower stratosphere accounting for a significant fraction of the chemical
831    ozone loss.   Bromine catalysis would account for nearly half of the total photochemical
832    sink of $O_3$ between 14 and 18 km in the mid-latitude stratosphere if the abundance of $Br_y$
833    is indeed 3 pptv at the tropical tropopause (*Figure 4.2*).
834
835    4.2.1 Testing strategy:
836
837    Measurements of bromine monoxide and the largest possible suite of precursors (e.g.
838    Halons, $CH_3Br$, $CH_2Br_2$, $CHBr_3$, etc.) in the region of the tropical tropopause will allow
839    closure of the stratospheric bromine budget. Because short-lived compounds are likely
840    involved, a single profile in the tropics will not be adequate.   Observations must be
841    obtained in various geographical regions and during several seasons to assure
842    representative sampling. Observations across the subtropical jet will enable better
843    understanding of bromine in the lowermost stratosphere.   Measurements of BrO in the
844    upper troposphere with a precision of at least 1 pptv are necessary to constrain the direct
845    input of $Br_y$ to the stratosphere. Aura MLS, SAGE-III, and SCIAMACHY limb
846    observations of BrO can be both validated and used to extend this analysis to the global
847    scale.
848
849    *4.3 Hypothesis* - **Iodine and stratospheric ozone:**
850
851       *Iodine monoxide is an important catalyst for the destruction of stratospheric ozone.*
852
853    Solomon *et al.*  (1994) suggested that iodine monoxide (IO) may be an important free
854    radical in the lower stratosphere. Since there are no long-lived iodine containing
855    compounds, any iodine reaching the stratosphere must be carried by short lived organic
856    and inorganic compounds or possibly on aerosol. The catalytic activity of iodine is due
857    mostly to the reactions of IO with $HO_2$.  Because this reaction rate is 3-4 times faster than
858    the corresponding rate of BrO, 1 pptv of IO contributes nearly as much as 3 pptv of BrO
859    to the loss rate of ozone in the lower stratosphere.
860
861    The stratospheric burden of iodine remains uncertain. Column observations by Wennberg
862    *et al.* (1997), illustrated in *Figure 4.3*, suggested that about 0.1 pptv of IO was present in
863    the lower stratosphere (assuming a column weighted near 120 mb).   Pundt *et al.* (1998)
864    obtained an upper limit of 0.1 pptv of IO at 15 km from balloon-borne observations.
865    Recently, however, Wittrock *et al.* (2000) have observed significant column abundances

of IO in ground-based observations obtained at high latitude. They attribute this signal to a stratospheric mixing ratio of IO equal to 0.65 to 0.8 pptv (*Figure 4.4*). Consistent with this result, K. Pfeilsticker's group have observed IO in solar transmission spectra obtained by a balloon-borne instrument at float (30-40 km). The retrievals are not yet complete but suggest IO densities similar to Wittrock *et al.* (K. Pfeilsticker, *personal communication,* 2000). It remains unclear whether the different results obtained by these researchers represent errors in the earlier measurements of Wennberg *et al.* and Pundt *et al.* or uncertainty in the photochemistry of stratospheric iodine. Recently, Bedjanian *et al.* (1998) have shown that OIO is produced with nearly unit efficiency in the reaction of BrO with IO. Depending on the photochemistry of OIO, this result may significantly alter the fraction of iodine present as IO. If the photolysis of OIO is slow, as suggested by Cox *et al.* (1999) and partially confirmed by Crowley *et al.* (2000), the fraction of iodine present as IO in the lower stratosphere may be considerably smaller than earlier models suggested (Solomon *et al.*, 1994; Wennberg *et al.,* 1997). This fraction may also vary significantly depending on the chemical sinks of this compound (with ClO for example).

*4.3.1 Testing strategy*:

Measurements of iodine monoxide and the largest possible suite of precursors (e.g., $CH_3I$, $CH_2I_2$, $C_2H_5I$, $CF_3I$) in the region of the tropical tropopause will allow closure of the overworld iodine budget. Because short lived compounds are likely involved, a single profile in the tropics will not be adequate. Observations must be obtained in various geographical regions and during several seasons to assure representative sampling. Observations across the subtropical jet will enable better understanding of iodine in the lower most stratosphere. SAGE-III and SCIAMACHY limb observations of IO can be both validated and used to extend this analysis to the global scale.

*4.4 Hypothesis* - **PAN, Oxygenated Hydrocarbons, and O$_3$ production in the UT**:

> *Oxygenated VOC photolysis both increases HO$_x$ levels and promotes the formation of PAN in the upper troposphere. The enhanced PAN formation significantly reduces the NO$_x$ / NO$_y$ ratio. This reduces the ozone formation rate in the upper troposphere and increases the input of NO$_y$ to the stratosphere.*

Singh *et al.* (1995) reported the discovery of high concentrations of oxygenated hydro-carbons in the remote troposphere. One of these compounds, acetone, is expected to significantly influence the photochemistry of the upper tropical troposphere (McKeen *et al.*, 1997). In particular, the photolysis of acetone is a source of odd-hydrogen radicals that can dwarf the traditional $O_3$/$H_2O$ source (*Figure 4.5*) (Singh *et al.*, 1995; Wennberg *et al.,* 1997). Coincident HO$_x$, acetone, and peroxide measurements have never been obtained in the tropical upper troposphere. Because the lifetime of acetone is quite short (10 days), extrapolation of the observations obtained in mid latitudes to the TTL is fraught with uncertainty.

911 In addition to the production of $HO_x$ radicals, acetone and other higher aldehydes produce
912 the acetyl radical upon photolysis. Peroxyacetyl nitrate (PAN) formation will occur in
913 the presence of $NO_2$. The formation of PAN has been suggested to significantly reduce
914 the $NO_x$ levels in the UT (Keim *et al.*, 1999; Folkins *et al.*, 2000). PAN is not soluble
915 and has a calculated photochemical lifetime in the UT/LS of 1 to 2 months and therefore
916 will likely be efficiently transported across the tropical tropopause. Observations of $NO_x$
917 and $NO_y$ in the tropics are quite limited. During ASHOE/MAESA and STRAT, $NO_y$ was
918 measured to be ~200-400 pptv at the tropical tropopause. Typically, NO comprised 30-
919 60% of the budget of $NO_y$.
920
921 *4.4.1 Testing strategy*:
922
923 Measurements of NO, $NO_2$, $NO_y$, $HNO_3$, $HNO_4$, PAN, acetone, and other oxygenated
924 organic compounds in the TTL will provide a critical test of this hypothesis. Because the
925 tropospheric sources of these compounds are inhomogeneous, a single profile in the
926 tropics will not be adequate. Observations must be obtained in various geographical
927 regions and during several seasons to assure representative sampling. TES and HIRDLS
928 are expected to retrieve many of these compounds (e.g. $NO_2$, $HNO_3$, PAN) and the
929 aircraft observations can provide validation of the retrievals. AURA and other satellite
930 observations of these compounds can be used to extend the higher spatial resolution *in*
931 *situ* observations to the global scale.
932
933 *4.5 Hypothesis* - **The stratospheric sulfate aerosol layer**:
934
935　　*Short-lived sulfur containing compounds contribute significantly to the background*
936　　*stratospheric aerosol.*
937
938 Understanding the mechanisms that determine the aerosol surface area in the lower
939 stratosphere during volcanically-quiescent times is critical for understanding the
940 interactions of the free radical families and therefore ozone. Recent studies have pointed
941 out that the OCS source is insufficient to explain the background stratospheric aerosol
942 (*e.g.,* Weisenstein *et al.*, 1997; Kjellstrom, 1998; Karcher and Solomon, 1999). Other
943 possible organic sources of stratospheric sulfur are $CS_2$, dimethyl sulfide (DMS), methyl
944 sulfonic acid (MSA). Observations of aerosol number and volume near the tropical
945 tropopause suggest significant aerosol mass is advected through the tropical tropopause
946 (Brock *et al.*, 1995). This result strongly suggests the existence of a TTL. In addition,
947 Murphy *et al.* (1998) have shown a veritable zoo of particle types in the tropopause
948 region. A 2D model calculation based on a recently determined upper limit for the UV
949 absorption cross section of $H_2SO_4$ significantly underestimates the sole atmospheric
950 profile of $SO_2$ obtained under quiescent conditions (*Figure 4.6*) (Burkholder *et al.,* 2000).
951 This comparison suggests the occurrence of an undetermined mechanism for conversion
952 of $H_2SO_4$ to $SO_2$ or perhaps the transport of significant amounts of $SO_2$ or organic sulfur
953 species other than OCS across the tropical tropopause. These studies point to unique and
954 interesting aspects of gas phase sulfur and sulfate aerosol chemistry that can be addressed
955 in the tropical upper troposphere and lower stratosphere. .
956

957 *4.5.1 Testing strategy*:

958

959 Measurements of aerosol properties (*e.g.*, particle size, composition, and the number of
960 condensation nuclei) as well as the abundance of both organic and inorganic precursors
961 of stratospheric sulfur in the TTL will provide a critical test of this hypothesis. The
962 measurements should be focused on improving our understanding of both the chemical
963 and physical processes responsible for aerosol formation in the stratosphere. Profiles of
964 OCS, $H_2SO_4$, and $SO_2$ can be measured by the balloon–borne MkIV spectrometer and
965 would be most useful if observed in the tropics. AURA and other satellite observations
966 of aerosol extinction at numerous wavelengths will provide global physical and perhaps
967 chemical characteristics of stratospheric aerosols. The proposed mission would provide
968 validation for these aerosol measurements near the tropical tropopause. The Aura
969 platform will only be able to measure profiles of $SO_2$ under volcanic conditions. If a
970 major eruption were to occur soon after launch of EOS Aura, a flight of MkIV would
971 provide important validation for profiles of $SO_2$, focused of course on a different
972 "hypothesis" than offered here.

973

974 # 5. Tropical Upper Tropospheric Water Vapor

975

976 5.1 *Introduction*

977

978 The response of the hydrological cycle to greenhouse gas induced climate forcing is
979 perhaps the single most important source of uncertainty in predicting future changes to
980 Earth's climate and composition. The small amounts of water vapor in the upper
981 troposphere (UT) exert enormous leverage on the radiative balance and composition of
982 the lower atmosphere. Increases in upper tropospheric $H_2O$ within global climate models
983 result in much greater radiative effects at the surface than are caused solely by the build-
984 up of $CO_2$ and other greenhouse gases (*e.g.*, Shine and Sinha, 1991; Ramanathan and
985 Collins, 1991).

986

987 Of particular importance is the moisture in the dry subtropical regions, which has a large
988 cooling effect on the whole tropics. As the radiation budget is more sensitive to
989 fluctuations for very dry air than for very moist air, changes in subtropical water vapor
990 could have a profound effect on Earth's climate (Spencer and Braswell, 1997). It is
991 therefore important to understand what processes control the subtropical humidity and
992 how climate change will affect these processes. These are key issues in determining the
993 nature of the water vapor feedback on climate.

994

995 The standard picture of the tropical troposphere is of large areas of gentle downwelling
996 punctuated by isolated convective regions of extremely rapid ascent, which make up only
997 a small fraction of the total area (only around a third of tropics is subject to direct
998 moistening by nearby deep convection). The outflow from these convective regions is
999 largest around 200 hPa (base of TTL), and as this air subsides the water vapor mixing
1000 ratio relaxes to very low values (10 ppmv or less). Observations show that the subtropics
1001 are dry, but not as dry as this simple picture would imply (*e.g.*, Pierrehumbert, 1998).

1002 Hence there must be an additional moisture sources that hydrate the regions of the tropics
1003 characterized by descent.
1004
1005
1006 *5.2 Hypotheses for supply of moisture to the subtropical free troposphere*:
1007
1008 1. **Evaporation of precipitation**:
1009
1010    *Evaporation of hydrometeors provides the main moisture source for the subtropics*
1011    *(Sun and Lindzen, 1993).*
1012
1013 2. **Evaporation of detrained cloud particles**:
1014
1015    *Evaporation of dissipating clouds provides the main moisture source for the*
1016    *subtropics.*
1017
1018 **3.  Lateral Transport**
1019
1020    *Outflow from tropical convective regions occurs over a deep layer (i.e. not restricted*
1021    *to altitudes above 200 hPa) and lateral transport by the large-scale wind field over*
1022    *this layer provides the additional supply of moisture* (Pierrehumbert, 2000 and
1023    *references therein).*
1024
1025 Which of the above hypotheses are correct, or more realistically the relative contribution
1026 of the three sources to subtropical moisture, has major implications on how subtropical
1027 humidity will change in response to climate warming (and hence the water vapor
1028 feedback on climate) (*e.g.*, Held and Soden, 2000; Pierrehumbert, 2000). This also has
1029 implications on requirement for climate models to accurately simulate tropospheric water
1030 vapor and water vapor feedbacks, *i.e.* the first two hypotheses require accurate
1031 representation of microphysical processes, whereas for the third hypothesis accurate
1032 representation of large-scale winds and in particular transient wave activity is most
1033 important (*e.g.*, Pierrehumbert and Roca, 1998).  For the **Lateral Transport** hypothesis,
1034 it is also important to establish from what level(s) in the atmosphere the advection of $H_2O$
1035 occurs.
1036
1037 There is compelling evidence that lateral transport is a significant source for subtropical
1038 moisture. In particular, simulations of water vapor distribution using analyzed winds are
1039 generally in good agreement with observations suggesting that the tropical humidity
1040 distribution in the free troposphere can be understood (at least to first order) by following
1041 air parcel trajectories backwards to their "temperature of last saturation" (*e.g.*, Sherwood
1042 1996; Salathe and Hartmann, 1997; Pierrehumbert, 1998; Pierrehumbert and Roca, 1998;
1043 Dessler and Sherwood, 2000). These simulations indicate that upper tropospheric water
1044 vapor distribution will contain many fine-scale features, with moist and dry plumes
1045 intertwined, and it has been suggested that the northern subtropics can be viewed as
1046 background of dry air interrupted by a few moist plumes (Pierrehumbert, 1998), see

*Figure 5.1*. Note that fine-scales exist not only in the horizontal, but also in the vertical due to differential advection that creates thin layers as in the stratospheric surf zone.

*5.2.1 Testing strategy*:

TC[3] has three major foci for testing the hypotheses of UT water vapor:

> A. confirmation of the accuracy of the AURA (and other satellite) measurements of $H_2O$ and MLS measurements of cirrus ice in the tropical and subtropical UT;

> B. determination of the horizontal and vertical scale of structures in UT $H_2O$ field;

> C. measurements of the isotopic composition of $H_2O$, together with measurements of tracers with a range of lifetimes, that will help determine the origin of upper tropospheric water vapor (and the relative contribution of above hypotheses to subtropical moisture budget).

Given the importance of water vapor in the radiative budget it is crucial to accurately know the water vapor distribution. Because of lack of data, however, there is currently uncertainty regarding the actual abundance of water vapor in the UT. Measurements from AURA (and other future satellites) will provide measurements of $H_2O$ in this region, and will greatly improve the situation. However, aircraft measurements in both moist and dry regions are required for validation of the satellite observations of $H_2O$.

The MLS measurement of cirrus ice will provide important constraints on the hypotheses for supply of moisture to the subtropical free troposphere. Development of a new in situ capability for measurement of condensed phase $H_2O$ should be given high priority both for validation of the MLS measurement and for testing these hypotheses on spatial scales (e.g., high vertical resolution) that are inaccessible to the satellite.

Addressing focus B will be an important part of the satellite validation. Because fine-scale structures are ubiquitous in UT $H_2O$ this variability will need to be considered when validating satellite measurements (with aircraft and other measurements), and also when interpreting the satellite measurements (in particular, measurements near the tropopause). Focus B is also important determining the processes that control subtropical moisture. In particular, if fine-scale features are not ubiquitous then this will cast doubt on lateral mixing being the dominant source of moisture in the subtropics as well as the TTL paradigm.

Each of the scenarios listed above (Hypotheses 1-3) will leave a distinct isotopic signature in $H_2O$ found in the dry downwelling regions. Focus C, the measurement of the isotopic composition of $H_2O$, will provide important new constraints on the origin of upper tropospheric water vapor, and will be vital for distinguishing between different mechanisms for controlling the moisture in each region. The isotopic composition of water vapor preserves a record of the condensation history of an air mass (see section 3.4), and profiles of $H_2O$, HDO, and $H_2^{18}O$ can be used to quantify the role of the various

sources of water vapor for the tropical and mid-latitude upper troposphere (Webster *et al*., 2000).

The signature for hypothesis 1, **Evaporation of Precipitation**, will be a relatively constant vertical profile of vapor isotopic composition. The lofted ice particles that supply water to this region will preserve the much heavier isotopic composition of their original condensation level. If these ice particles are the primary source of water, the ambient levels of HDO and $H_2^{18}O$ will reflect primarily the composition of the particles, with little fractionation upon evaporation. The signature for hypotheses 2 and 3 will be a sloped profile of vapor isotopic composition that becomes lighter with increasing altitude. For hypothesis 3, **Lateral Transport**, the isotopic ratios should act as "conserved tracers" as air parcels from convective and downwelling regions mix. At a specific potential temperature, correlation plots of HDO vs $H_2O$ and of $H_2^{18}O$ vs $H_2O$ should define "mixing lines" that can be used to quantify the relative contribution of each air mass type to the resulting admixture. In regions where profiles of HDO and $H_2^{18}O$ vs altitude become progressively lighter with increasing altitude, the failure to observe such mixing lines will suggest that hypothesis 2, **Evaporation of Detrained Cloud Particles**, is occurring. Comparison of plots of HDO vs $H_2^{18}O$ can yield information about mixing of air masses from different altitudes. Therefore, measurements of the isotopic composition of $H_2O$ may provide quantitative information regarding the atmospheric level(s) from which water is advected according to the **Lateral Transport** scenario.

To address all the above issues aircraft measurements will be required in both (tropical) moist and (subtropical) dry regions, and key flights will be those that cross between these two types of regions (i.e., flights that cross strong gradients in water vapor). It will also be important (for validation of the lateral mixing hypothesis) to flight within regions where transport between moist and dry regions is predicted to occur. This can be done by the same series of flights proposed below.

## 6. Observational Strategies and Instrument Development

*6.1 Observational Strategies for TC$^3$*

The Tropical Composition and Climate Coupling Experiment will combine AURA and other satellite measurements with in situ and remote observations obtained from a high altitude aircraft (the ER-2 and/or WB-57) deployed from two Pacific sites: Guam (13.4°N, 144.4°E) and Hawaii (21.3°N, 157.9°W). NASA aircraft have operated out of both of these sites and transit between the sites can be made in single flights of either the ER-2 or WB-57. These sites will enable observations of the two distinct meteorological regions - areas of active convection and areas of downwelling on both sides of the ITCZ. The following information is based on climatological fields of wind, temperature, outgoing longwave radation (OLR), upper troposphere $H_2O$ from MLS, and lower stratosphere $H_2O$ from HALOE shown in *Figures 2.2* and *2.3*. (See also **http://code916.gsfc.nasa.gov/Personnel/people.Newman,_Paul_A./easton/easton.html** for more information on these climatologies).

1138
1139    During Northern Winter:
1140
1141    Guam is in a non-convective region (low clouds, high OLR), but to the south (and within
1142    reach by the ER-2 or WB-57) is a region of strong convection. There is a strong north-
1143    south gradient in UT water vapor over Guam (*Figure 2.2*), with high water vapor to the
1144    south over the region of strong convection. Within the neighborhood of Guam there are
1145    also important variations of temperature and $H_2O$ near the tropopause. The cold pool at
1146    100 hPa is centered just about over Guam, while (according to HALOE) the region of
1147    minimum water vapor at the tropopause is slightly north of Guam. There are also strong
1148    variations in the horizontal flow, in both the UT and LS (i.e. 200 to 70 hPa), near Guam,
1149    which are possibly important for both stratospheric dehydration and redistribution of UT
1150    moisture.
1151
1152    Therefore a NH winter deployment with principal base in Guam, with flights to the south
1153    and the north, would sample
1154
1155        • convective and non-convective regions
1156
1157        • high and low UT water vapor (and cross steep gradients in UT $H_2O$)
1158
1159        • the cold pool in the tropopause or TTL
1160
1161        • near the tropopause the region of minimum $H_2O$.
1162
1163    The conditions in the region around Hawaii are very different from those over Guam
1164    during northern winter, and flights from Hawaii would enable an even larger contrast
1165    than the Guam-based flights. Sampling lower UT water vapor and tropopause air away
1166    from the cold pool as well as the region where large lateral transport is expected
1167    (transport from extratropics into tropics both above and below the tropopause, and
1168    transport moist tropical UT into dry subtropics). The Hawaii component of the
1169    deployment may only need to consist of 1 flight into Hawaii from Dryden, 1 flight due
1170    south and back, and 1 flight to Guam, with reverse for flights back to Dryden.
1171
1172    During Northern Summer:
1173
1174    A corresponding NH summer deployment would provide valuable information on
1175    seasonal variations in chemistry, transport, and $H_2O$. As shown in *Figure 2.3*, the
1176    conditions in the summer are very different than winter Summer flights would enable
1177    investigation of the influence of the Asian monsoon on the composition of the tropical
1178    UT/LS (from Hawaii it is possible to reach the preferred region for exchange across the
1179    northern subtropical tropopause, *e.g.*, Figure 4 of Postel and Hitchman 1999) and may
1180    provide key information for understanding the cause of the seasonal cycle and apparent
1181    long-term rise of stratospheric $H_2O$.
1182
1183    Notes:

1184
1185 - Need to determine whether it is possible to sample "outflow" of Asian monsoon from
1186 Guam/Hawaii, both in UT and LS.

1187
1188 - For possible moistening of tropical stratosphere, Sep-Oct may be preferred season.

1189
1190 - Need to talk a bit about the balloon deployment (s) from the tropics: will the site used
1191 previously in Brazil be good enough?

1192
1193 ***Finally: it is beyond the scope of this draft white paper to delve into which high altitude***
1194 ***aircraft is preferable. Clearly there are advantages and disadvantages for each.***
1195 ***Ideally, the mission will involve both high-altitude aircraft as well as a lower-altitude***
1196 ***aircraft that will provide LIDAR measurements and in situ observations of the***
1197 ***composition of air that is convected to the upper troposphere. Aircraft issues, as well***
1198 ***as details of the payload(s), are better handled after a consensus has been reached on***
1199 ***the scientific goals of a tropical mission.***

1200
1201
1202
1203 *6.2 New Instrumentation Needs for TC$^3$*

1204
1205 The previous sections have outlined a number of new instrument needs that will be
1206 required to test the various hypotheses. All of these measurements must be obtained at a
1207 vertical resolution of order 100 m (e.g., *high temporal resolution sampling is required*) to
1208 test the TTL paradigm and the associated "hypotheses of stratospheric aridity",
1209 "hypothesis of LS and UT photochemistry", and "hypotheses of UT water vapor". It is
1210 conceivable that some of these measurement requirements could be met by a Whole Air
1211 Sampler system. If that is the case, this system must be capable of obtaining more
1212 samples than are returned by the current ER-2 system. The new measurement needs are:

1213
1214 • Water Isotopes (both HDO and $H_2O^{18}$)
1215 • Condensed Phase $H_2O$
1216 • Short Lived Tracers of Convection (Ra, $^{210}Pb$, $^{222}Rn$, $CH_3I$)
1217 • Oxygenated Hydrocarbons (PAN, acetone), $H_2O_2$, and possibly $HNO_4$
1218 • Short Lived Organic Bromine Source Species ($CH_3Br$, $CHBr_3$, $CH_2Br_2$, Halons,
1219   etc)
1220 • Iodine Monoxide (IO) and its primary Organic Source ($CH_3I$). Measurements of
1221   $CH_2I_2$, $C_2H_5I$, $CF_3I$ are also desired.
1222 • Sulfur Dioxide ($SO_2$) (from an airborne platform) and its organic sources $CS_2$,
1223   dimethyl sulfide (DMS), methyl sulfonic acid (MSA).

1224

# 7. Summary of Hypotheses for TC[3]

The following hypotheses will be directly tested by the proposed tropical high-altitude aircraft flights from Guam and Hawaii, balloon flights from an as yet undetermined tropical site(s), and measurements from the AIRS, HIRDLS, MLS, and OMI instruments on board EOS Aura. An evaluation of these hypotheses will play a *central role in gaining quantitative understanding of the couplings between atmospheric composition and climate*:

**There is a tropopause transition layer (TTL) in the tropics between 14 and 20 km charcacterized by**:
    **slow ascent** (driven by extratropical wave driving in the stratosphere) and quasi-horizontal mixing with the extratropics;
    **lateral mixing** into/out of/within this layer is limited to certain regions and seasons: downstream of the summer monsoons, and within the northern winter westerly ducts over the Pacific and Atlantic oceans.

**Stratospheric Aridity is maintained either by:**
    Class I Hypotheses (One dimensional models):
        1) **Cold Trap** - slow, large-scale upwelling through the cold tropopause dehydrates ascending air
            1a) *Fountain* - air enters the stratosphere only in the Western Pacific.
        2) **Convective Overshoot** - Convective turrets ascend rapidly from near the ground and penetrate into the stratosphere, where they induce rapid dehydration due to precipitation of ice .
            2a) *Ice Lofting* - air entering the stratosphere contains significant amounts of ice, some of which precipitates and some of which evaporates.
            2b) *Supersaturation* - air entering the stratosphere is highly supersaturated (*e.g.*, 150%) with respect to cloud top temperature
    Class II Hypotheses (Three dimensional models):
        3) **TTL Cold Trap** - air enters the TTL to the west of the region of coldest temperatures in an area of forced upwelling and parcels are advected to the region of coldest TTL temperatures by an anti-cyclonic flow. As the air approaches the region of coldest temperature, it sinks, cools, and desiccates over relatively long time scales (several days).
        4) **TTL Overshoot** - convective turrets penetrate the TTL. The more energetic turrets penetrate to higher levels and become rapidly (time scales of less than a day) dehydrated due to sedimentation of ice. The resulting humidity of the TTL is caused by mixtures of air parcels that detrain at different potential temperatures.
        5) **Stratospheric Dehydration** - Dehydration in the lowermost stratosphere is induced by meso or synoptic-scale wave activity that lifts and cools air masses in the lower.

**Sources of stratospheric bromine:**
    Short-lived bromine containing compounds enhance significantly the concentration of bromine monoxide in the upper troposphere and lower stratosphere. As a result, ozone loss rates in the lower stratosphere are significantly faster than expected.

**Iodine and stratospheric ozone:**
    Iodine monoxide is an important catalyst for the destruction of stratospheric ozone.

**PAN, oxygenated hydrocarbons, and O$_3$ production in the UT:**

    Oxygenated VOC photolysis both increases HO$_x$ levels and promotes the formation of PAN in the upper troposphere. The enhanced PAN formation significantly reduces the NO$_x$/NO$_y$ ratio. This reduces the ozone formation rate in the upper troposphere and increases the input of NO$_y$ to the stratosphere.

**The stratospheric sulfate aerosol layer**:

    Short-lived sulfur containing compounds contribute significantly to the background stratospheric aerosol.

**The supply of moisture to the subtropical free troposphere is maintained either by:**

    **1. Evaporation of precipitation**: Evaporation of hydrometeors provides the main moisture source for the subtropics.
    **2. Evaporation of detrained cloud particles:** Evaporation of dissipating clouds provides the main moisture source for the subtropics.
    **3. Lateral Transport:** Outflow from tropical convective regions occurs over a deep layer (i.e. not restricted to altitudes above 200 hPa) and lateral transport by the large-scale wind field over this layer provides the additional supply of moisture.

Finally, the study of stratospheric aridity is motivated in part by the need to understand the cause of the apparent rise in stratospheric H$_2$O during the 1990s and to clearly define future changes to stratospheric H$_2$O that occur during the EOS Aura era. Our mission is focused on elucidating the mechanism(s) that govern stratospheric H$_2$O and on validating a suite of measurements to be obtained from EOS Aura. Testing the hypotheses of stratospheric aridity will provide insight into the climatic variables (e.g., surface temperature; tropopause temperature; convective energy, etc.) that affect stratospheric H$_2$O (*e.g.*, Section 3.2). Our proposal to conduct this mission over several seasons, and several years, is designed to provide additional insight into how seasonal and/or interannual variability in processes in the hypothesized tropical TTL affect stratospheric H$_2$O. In this way, the *TC$^3$ mission will lay the ground-work for the use of Aura satellite observations to test the following hypotheses*:

**An increase in stratospheric H$_2$O may be caused either by:**

    **Rising Cold Trap Temperature** - Stratospheric H$_2$O responds directly to warming of the tropopause region.
    **Increasing Asian Monsoon** - An increase in the extremity of the Asian monsoon over Indonesia leads to a moister upper troposphere, resulting in greater abundance of H$_2$O within the TTL for air parcels that do not pass directly through a cold trap.
    **Changing Ice Physics** - Convective turrets become less efficiently dehydrated over time due to subtle changes in the microphysics of ice.
    **Reduced Brewer-Dobson Circulation** - Stratospheric wave activity in the NH is reduced, resulting in increased fraction of air entering the TTL during NH.

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## 9. List of Figures

The figures referred to in this white paper are saved as individual files on the server containing this document. Captions are included with each figure. Both postscript (*.ps) and "portable document format" (*.pdf) files are provided for each figure. Please contact one of the authors if you are having difficulty accessing or printing the figures. The figures are numbered according to the section of the document where they first appear. The list of figures is: